Paper

^{210}Po IMPLANTED IN GLASS SURFACES BY LONG TERM EXPOSURE TO INDOOR RADON

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Abstract—**Recent epidemiologic investigations of the relationship between residential radon gas exposure and lung cancer relied on contemporary radon gas measurements to estimate past radon gas exposures. Significant uncertainties in these exposure estimates can arise from year-to-year variation of indoor radon concentrations and subject mobility. Surface implanted ^{210}Po has shown potential for improving retrospective radon gas exposure estimates. However, in previous studies, the ability of implanted ^{210}Po activity to reconstruct cumulative radon gas exposure was not tested because glass was not available from homes with known radon-gas concentration histories. In this study, we tested the validity of the retrospective radon gas reconstruction using implanted ^{210}Po surface activity by measuring glass surfaces from homes whose annual-average radon gas concentrations had been measured almost every year during two decades. Regression analysis showed a higher correlation between measured surface activity and cumulative radon gas exposure in these homes ($R^2>0.8$) than was observed in homes where only contemporary radon gas measurements were available. The regression slope (0.57 ky m^{-1}) was consistent with our earlier retrospective results. Surface activity measurements were as reliable for retrospective radon *gas* exposure reconstruction as yearlong gas measurements. Both methods produced estimates that were within 25% of the long-term average radon gas concentrations in a home. Surface measurements can be used for home screening tests because they can provide rapid, reliable estimates of past radon gas concentrations. Implanted ^{210}Po measurements are also useful in retrospective epidemiologic studies that include participants who may have been exposed to highly variable radon concentrations in previously occupied or structurally modified homes.**

Health Phys. 83(2):261–271; 2002

Key words: radon progeny; exposure, radiation; epidemiology; monitoring, air

INTRODUCTION

The National Research Council's BEIR VI report estimates that approximately 18,600 lung cancer deaths in the U.S. population each year may be caused from residential exposure to ^{222}Rn gas (radon) decay products (NRC 1999). Case-control epidemiologic investigations have attempted to examine the relationship between cumulative radon gas exposure and lung cancer. Most studies used one or more contemporary radon gas concentration measurements that lasted for 1 y or less. The cumulative radon gas exposure was estimated to be the product of the short-term contemporary radon gas concentration in the house, adjusted to yearly average, times the years spent in that house, and summed across houses occupied during the 20 to 25 y prior to measurement. More advanced studies take the individuals' mobility and occupancy into account to adjust for the different exposure times spent in different radon concentrations. Contemporary radon gas measurements can introduce errors in total dose estimates because residential radon gas concentrations vary from year-to-year (Steck et al.1990; Martz et al. 1991; Steck 1992; Swedjemark et al. 1994) even in homes without structural or occupant changes.** Additional errors are introduced if the study subjects occupied more than one house during the reconstructed period (Field et al. 1996). Previous residential radon case-control epidemiologic studies have had to impute from 17–40% of their radon exposures for time spent by the study participants in various dwellings where radon measurements were unavailable. The missing data create significant gaps in the participants' exposure history, which compel the investigators either to analyze a reduced data set or to impute radon concentrations for missing homes (Weinberg et al. 1996). These gaps in radon measurements seriously decrease a study's statistical power to detect an association, especially if the gaps occur 5 to 15 years prior to study enrollment (NRC 1999; Lubin et al. 1990).

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(*Manuscript received* 11 *October* 2001; *revised manuscript received* 18 *January* 2002, *accepted* 19 *February* 2002)

0017-9078/02/0

** Steck DJ. Variations of annual average 222Rn concentrations in homes over two decades. Health Phys (in preparation 2002).

Surface implanted ^{210}Pb has been investigated as a means of overcoming the exposure assessment obstacles noted above (Lively and Ney 1987; Samuelsson 1988; Steck et al. 1990; Samuelsson et al. 1992; Lively and Steck 1993; Mahaffey et al. 1993; Steck et al. 1993; Falk et al. 1996; Mahaffey et al. 1999; Steck and Field 1999a and b; McLaughlin 2001; Falk et al. 2001). A glass surface exposed to radon gas over an extended period of time will develop a surface radioactivity that depends on the product of the radon gas concentration and the exposure time of the surface. A fraction of the airborne, short-lived alpha emitting radon gas decay products deposit on room surfaces and can implant nuclei, including ^{210}Pb, in glass surfaces in a room. The fraction that is deposited also depends upon environmental factors like air movement and aerosols. Glass that is not covered by a porous hydrate (Fleischer and Doremus 2001) provides a stable matrix for the ^{210}Pb deposit. The 22-y half-life of ^{210}Pb means that the activity grows slowly towards equilibrium and persists for decades after the exposure ends. This predictable temporal behavior provides a long-lasting marker for past radon concentrations. The alpha particles emitted from ^{210}Po, a ^{210}Pb decay product, provide the easiest measure of the trapped surface activity. Etched track detectors are well suited to measuring the alpha activity in glass surfaces. The alpha particles from the glass produce tracks in the detector at a rate that is proportional to the alpha activity in the glass, which, in turn, depends on the cumulative radon gas exposure. Once a calibration is established between the track generation rate in the detector and the cumulative radon gas exposure, then the average radon gas concentration during the glass' exposure can be determined. Glass that has remained at a single location can be used to establish the cumulative radon gas exposure in that space while glass that has been carried with the individual from home to home can be used to estimate the cumulative radon gas exposure in those spaces after appropriate adjustments have been made for the activity's temporal response.

Previous laboratory work on this technique established that an excellent correlation exists between the measured cumulative radon gas exposure and the implanted ^{210}Po activity on glass surfaces (Lively and Steck 1993). In previous field studies, where the cumulative radon exposures in homes were estimated from contemporary radon gas measurements, the correlation between radon gas in homes and implanted activity was lower but encouraging (Samuelsson 1988; Lively and Steck 1993; Mahaffey et al. 1993; Steck et al. 1993; Falk et al. 1996; Mahaffey et al. 1999; Steck and Field 1999a and b; Falk et al. 2001; Birovljev et al. 2001). Temporal variability of radon concentrations, differences in deposition conditions, and differential leaching of ^{210}Po can reduce the correlation between estimated cumulative exposure and implanted activity. Improved correlation between radon exposure and implanted activity are possible when adjustments are made for the deposition environment of the surface (Steck and Field 1999a and b; Fitzgerald and Hopke 2000; Walsh and McLaughlin 2001). However, it is difficult to assess the accuracy of the glass technique from these studies as they have limitations such as small sample sizes, short exposure times, or limited measurements of the true radon exposure of the glass.

The goals of this study were to

1. compare surface activity measurements vs. contemporary radon gas measurements in their ability to reconstruct cumulative radon gas exposures in homes with known radon gas histories;
2. compare the relative performance of two different surface activity detectors; and
3. examine the reliability of the retrospective radon gas exposure reconstruction from surface implanted ^{210}Po in different deposition environments.

METHODS

Radon gas concentration measurements

Year-long radon gas measurements were made in 24 single-family houses and 1 college office building starting in 1983 (Steck 1990). These homes were selected from a larger group of houses that were participating in a long-term radon study (Steck 1992). All but two of the houses had basements that served as living spaces. The willingness of the homeowner to participate, the completeness of the radon measurement history, and the availability of suitable surfaces for glass measurements were selection criteria for inclusion in the glass study. Science teachers, college alumni, or their neighbors owned the houses. The adult occupants remained the same throughout the survey.

In each house, annual average radon concentrations were measured at two or more locations, usually on the two lowest levels, during the period 1983–1999. Each October, an alpha track detector (ATD) was placed for a yearlong exposure. Most homes have one or more measurements in the early 1980's and continuous measurements from 1990 to 1999. One home (site SD) has continuous measurements that began 1 y after the house was constructed in 1981. The radon gas measurements were made with alpha track detectors that have a 2-cm^2 chip of dosimetry grade CR-39 enclosed in a small plastic chamber. The chamber's interior was separated from room air by a convoluted, labyrinthine diffusion path to eliminate thoron and directly deposited airborne radon progeny. The chips were developed for 6 h in 6.25N NaOH and read under 100× magnification until a

minimum of 150 tracks were found. This method gives a reading reproducibility of approximately 10%.

Each batch of track registration material underwent an annual calibration test to identify potential changes in efficiency. This calibration took place in a large volume radon chamber monitored by calibrated continuous radon monitors. The calibration was validated, almost every year, by exposures in national radon chambers, usually as part of an international radon gas detector intercomparison exercise conducted by the U.S. DOE or the U.S. EPA. The detector was listed as meeting proficiency requirements as part of the U.S. EPA Radon Measurement Proficiency Program (Steck 2001). Adjustments to the calibration coefficient were made twice (1988 and 1990) when that batch of material showed a 10% loss of efficiency. An historical sample of detectors was reread in 1999 to judge the variation of the human reader's track-recognition reproducibility over the years. The total instrumental variability of the ATDs is estimated to be approximately 14% over the 17-y monitoring period based on the variability of the track material calibration and reader's historical reproducibility. Additional details on these ATDs are available elsewhere (Steck 1990, 1992).

Imputation for missing radon gas measurements

Missing annual radon gas measurements were imputed to obtain a cumulative gas exposure estimate that corresponded temporally to the cumulative surface activity measurements. Let A_0 = year in which glass object was acquired (first year in the time series being established) and A_i = year subsequent to A_0 ($i = 1 \ldots n$), where A_n is the year 2000.

Then P_i is an ordinal number indicating the number of the year subsequent to year:

$$A_0{:}P_i = A_i - A_0 + 1 (i = 1 \ldots n). \quad (1)$$

Let

$Q_{ik[j(m)]}$ = 0 if no radon gas measurements were made in year i ($i = 1 \ldots n$), house k ($k = 1 \ldots 80$), floor j ($j = 0 \ldots 3$) where 0 indicates a basement, and each house has three floors maximum above the basement, and replicate m ($m = 1,2,3$) = 1 if a radon gas measurement was made in year i, house k, floor l, and replicate m;

$Y_{ik[j(m)]}$ = radon gas concentration in Bq m^{-3} made in year i, house k, floor j, and replicate m; and

$\epsilon_{ik[j(m)]}$ = random error associated with the measurement of radon gas concentration made in year i, house k, floor j, and replicate m.

In this data set, no house k had more than one intervention (e.g., remediation that would affect radon levels) in year A_r ($A_r = 1 \ldots n$).

Let

$R_{ik1} = 1$ for $A_i < A_r$ or if no intervention occurred;
$= 0$ for $A_i \geq A_r$;
$R_{ik2} = 1$ for $A_i \geq A_r$;
$= 0$ for $A_i < A_r$.

A regression equation with parameters β_0, β_1, β_2, and β_3 was estimated for each $ik(j)$. These parameters were tested for significance from zero to determine if the effect of the intervention on the radon gas concentration in the house was significantly changed. The regression equation is

$$Y_{ik[j(m)]} = [\beta_{0ik} + \beta_{1k}P_iQ_{ik(j)}]R_{ik1} + [\beta_{2ik} + \beta_{3ik}P_iQ_{ik(j)}]R_{ik2} + \varepsilon_{ik[j(m)]}. \quad (2)$$

To estimate the most appropriate model, various tests of hypotheses were conducted to determine if the parameters for year, house, and floor were affected by the intervention. The resulting equation was used to predict multiple values of *Y-hat*, which were imputed into the time series of measured radon gas concentrations in air for years in which $Q_{ik[j(m)]} = 0$. The imputation replaced missing values with predicted values plus random residuals based on the mean-square error (MSE) from the regression. The strength of this approach is that the mean and variance of the distribution of radon gas measurements are unchanged, as are estimates that depend on the mean and variance (e.g., correlations, confidence intervals, α-levels, p-levels). Note that if there was no intervention in a home, this model reduces to a simple linear regression. Only one house had a statistically significant response to an intervention that required this special modeling.

In this data set, an average of 7.5 y of data were missing from an average of 18.6 y. Thus, only 40% of the annual gas concentrations of radon needed to be imputed. To achieve a statistical efficiency of 95%, eight sets of imputations were required. That is, for each object measured in each house, eight radon gas time series were created. Each was used in further analyses, and results were combined for overall estimation and tests of hypotheses. An estimate of radon gas exposure of the glass was created by summing yearly measurements (actual plus imputed) corresponding to the age of the glass object. There were eight estimates, one for each of the eight imputations. Call these Z_{pkqr}, where Z_{pqkr} = cumulative radon gas exposure in kBq y m^{-3} for object p in house k for data set q ($q = 1 \ldots 8$) corresponding to the age of the glass object measured using detector type r ($r = 1,2$).

Surface activity measurements

The two detectors compared in this paper have been used previously in major epidemiologic residential radon studies. The Missouri Radon Lung Cancer Study (Alavanja et al. 1999) and the Iowa Radon Lung Cancer Study (Field et al. 2000) were both case-control epidemiologic studies that evaluated the lung cancer risk posed by residential radon exposure. The Iowa and Missouri studies used both yearlong contemporary radon gas measurements and surface activity measurements to obtain separate estimates of historic radon concentration. The Missouri study inclusion criteria allowed subjects to have lived in more than one home over the 20 y prior to enrollment. That study included glass objects that had traveled from home to home with the subject. The Iowa study only enrolled subjects who had lived in the current home a minimum of 20 y and selected glass objects that had been in the house for many years.

Since the two surface activity detectors evaluated in this paper are similar in many ways but different in some key characteristics, we shall refer to them with similar but distinctive acronyms based on their previous names. The device used in the Missouri Radon Lung Cancer Study was developed at the Pacific Northwest National Laboratory in Richland, Washington, and was called the CR-39 Surface Monitor in early publications. It will be referred to as the Retrospective Surface Monitor (RSM) in this paper. The device used in the Iowa Radon Lung Cancer Study was developed at the Physics Department, St. John's University, Collegeville, Minnesota. It will be referred to as the Retrospective Reconstruction Detector (RRD) rather than its earlier name: the Historic Reconstruction Detector (HRD).

Both devices measure implanted ^{210}Po activity in glass surfaces using dosimetry grade track registration material. The RSM uses a 25-cm^2 piece of CR-39 plastic manufactured by American Technical Plastics, Inc. (Stratford, CT) while the RRD uses a 4-cm^2 piece of LANTRAK (Landauer, Inc., Glenwood, IL). Each device used protective coverings to reduce pre- and post-placement exposures. In this study, we used 3% lab blanks and 5% field blanks to monitor pre- and post-placement contamination. The RRD has a 0.8 mg cm^{-2} metalized Mylar cover between the detector and the glass to reduce UV damage and tracks from the natural alpha emitters present in the glass. The RSM is exposed without a cover but compares track morphology with a standard ^{210}Po-exposed detector to reduce the tracks from natural contaminants.

The glass surfaces were selected following the guidelines common to both RSM and RRD placement protocols (Mahaffey et al. 1993, 1999; Steck and Field 1999a). Glass surfaces were ordinary, smooth glass without visible coatings or colorings. The glass had been purchased by the homeowners and kept in the house during the time when the radon gas measurements were being made in that house (10 to 20 y). The preferred surfaces included interior-facing picture glass, mirrors, and door glass in unobstructed locations without strong air currents. Window glass surfaces were allowed; but south-facing window glasses were only used as a last resort for RSM placements in this study as these surfaces were not allowed in the Missouri study. One of the authors (DS) installed the detectors on alcohol-washed areas as near the center of the glass as the homeowner would allow. Data regarding the cleaning and smoking history in the house, room characteristics, and glass history were gathered by interviewing the homeowner during the installation visit. The detector pairs (1 RSM and 1 RRD) were placed as near as possible to the location where the radon gas measurements were made. Duplicate RSMs and RRDs were placed at 8% of the placement sites to examine the precision of the measurements. Approximately half of the detectors were returned by mail and the installer retrieved half. At 3% of the sites, RRDs and RSMs were placed on the same piece of glass immediately following the initial measurement for a sequential measurement.

The RSMs were exposed for 4 to 5 wk in the Missouri study. The RRDs were exposed for 1 y in the Iowa study. As a compromise, we exposed both devices for approximately 60 d during the present study. RRDs were placed for a yearlong exposure on 67% of the surfaces to test for any exposure time dependence. After exposure, both devices were chemically etched and manually read with a microscope. The area evaluated on the RSM varied from 0.1 to 0.2 cm^2 depending on the track density. The total tracks identified on each RSM normally ranged from 60 to 600. The RRD surface was examined in 0.025-cm^2-sized areas until a minimum of 150 tracks were identified. Unexposed detectors were analyzed to determine the number of blank tracks. The tracks from exposed detectors were converted to ^{210}Po activity density (Bq m^{-2}) by subtracting blank tracks, dividing by the area, the exposure time, and the detector efficiency. Eight percent of the detectors were exposed to calibrated, glass-implanted ^{210}Po standards. Both detector types showed similar efficiencies for ^{210}Po (RSM = 25%; RRD = 29%) based on their response to the standards. The ^{210}Po activity was age-adjusted to be equivalent to the activity that the implanted ^{210}Pb would have had, had there been no decay. This correction factor has a value of 1.4 for a 20-y-old surface. The naturally occurring alpha emitters in 21 glass samples created tracks in the RRD at an average rate that was equivalent to 1 Bq m^{-2}. This contamination rate was subtracted from the RRD results.

Additional details about the RSM and RRD are available elsewhere (Mahaffey et al. 1993, 1999; Alavanja et al. 1999; Field et al. 1999; Steck and Field 1999a and b).

Comparison of surface activity detectors

The surface activity densities of the two detectors should be linearly related since they share many common characteristics and protocols. Statistical analyses included the Pearson Product Moment Correlation Coefficient to test for differences between the surface activities of the RSM and RRD, and either a paired *t*-test (for normally distributed data) or a Wilcoxon Signed Rank Test (for data that could not be transformed to a normal distribution) to test for systematic bias between the track density rates. The Kolomogorov-Smirnov test was used to assess the normality of the data. Geometric means (GM) and geometric standard deviations (GSD) were used as summary descriptors for the field intercomparison results because of the lognormal nature of the track density data. Regression analyses were used to investigate the differences between the detectors' results and to examine the influence of specific high-leverage data and outliers. Hypotheses were tested to determine if the regression intercept was significantly different from zero and if the slope was significantly different from one.

Analysis of radon gas exposure vs. surface activity

The data from each glass object in each residence consisted of eight radon gas air concentration estimates and two surface activity measurements. Physical models of the deposition and implantation of nuclei in stable surfaces predict a proportional relationship between total radon gas exposure and implanted surface activity. The constant of proportionality depends on the deposition and implantation conditions. These environmental conditions may be different for each surface (Knutson 1988; Cornelis et al. 1992). However, we did not apply any environmental adjustments to the implanted activity in this analysis. Past comparisons suggest that the presence of tobacco smoke may be a major factor in the variability (Field et al. 1999). A constant term was included in the regression model to allow the fit to adjust for the possible effects of naturally occurring alpha emitters in the glass or activity loss. Eight identical analyses were conducted, corresponding to each of the eight data sets of radon gas concentration measurements. Let X_{pkqr} = age and decay corrected ^{210}Pb surface activity in Bq m^{-2} for object p in house k for data set q from surface alpha activity measurement method r and δ_{pkqr} = random error associated with the surface activity measurement on object p in house k for data set q by surface activity measurement method r.

Correlations were calculated with the original and log-transformed data including and excluding a home whose results gave it high leverage. Regressions were performed separately with RSM and RRD data using the following equation for each of the eight data sets, again including and excluding a high-leverage home:

$$Z_{pkqr} = \gamma_{0kqr} + \gamma_{1kqr}X_{pkqr} + \delta_{pkqr}. \quad (3)$$

By appropriately combining results across the eight data sets (Meng and Rubin 1992; Rubin 1987), hypotheses were tested to determine if γ_0 and γ_1 are significantly different from zero. The coefficient of determination (R^2) is used to judge the performance of the surface activity measurements to reconstruct the cumulative radon gas exposure.

Comparison of retrospective radon gas reconstruction methods

We used our data in a random sampling simulation to determine whether yearlong radon gas measurements with ATDs or surface activity measurements with RRDs or RSMs would yield results that are more representative of historical radon *gas* concentrations. We compared estimates of the cumulative radon exposure based on yearlong radon gas measurements and surface activity measurements against measured cumulative radon gas exposures through a repetitive sampling procedure. We tested 3 and 5 years sampling windows to mimic most epidemiologic studies' protocols. For each site, we randomly selected an actual radon gas measurement taken during 1 y of the multi-year measurement period and calculated the cumulative radon gas exposure as a product of the concentration and the age of the glass. Simultaneously, we randomly selected one of the eight cumulative radon exposure estimates based on the full set of radon gas measurements for each site (Z_{pqkr} as described above). We used the regression relationship to generate the surface activity's estimate of the cumulative radon exposure at each site. A linear regression was performed to obtain a coefficient of determination (R^2) for the contemporary radon gas and surface activity estimates to the cumulative exposure. This process was repeated eight times to obtain coefficients of determination distributions. The entire procedure was performed for two multi-year sampling periods to estimate the range of variation in the coefficients of determination caused by different distributions of missing yearlong measurements. To simulate common current practice in epidemiologic studies, we repeated the analyses using house-wide average radon gas and surface activity measurements instead of single site data.

RESULTS

Sixty-nine glass surfaces were measured with either a RRD (Iowa study) or a RSM (Missouri study) detector. The glasses were located in 51 different rooms of 24 different houses and 1 office building. We omitted one house (SD) from the summary statistics (Table 1) since it had an unusually large number of sample sites, 21 surfaces in 6 rooms. The median building age was 28 y, and the current occupant had been in the building for an average of 24 y. The distributions of the radon gas exposure and surface activities were lognormal if house SD was excluded, and more lognormal than normal, if it was included. We also performed subset analyses on the intercomparison data to see if house SD distorted the relationships.

Radon gas exposures

The average radon gas concentration in the study rooms was 177 Bq m^{-3}. Most houses showed no specific pattern of change from year-to-year. They tended to vary randomly about the long-term average by approximately 22% beyond the 14% instrumental uncertainty associated with the ATDs. These rooms do not represent a random sample as we avoided older homes that had been mitigated or modified and, hence, had a poorly known radon history. We did include two houses that had some active intervention but also had an extensive radon measurement history. We also favored easily accessible houses, those with elevated radon, and houses with current or ex-smokers in an attempt to study the effects of smoking on implantation.

The glass ages were log-normally distributed with a median age of 19 y and a range from 7 to 50 y old. Given the extensive radon measurement history, we had to impute only a small percentage of the glasses' cumulative radon exposure. We categorized the exposure data into two classes based on the product of the temporal variation percentage and the imputed exposure percentage. The exposure data clearly separated into two groups with approximately 85% in the well-characterized group. We used these categories to test the effects of imputation on the surface activity.

Table 1. Exposure conditions for glass surfaces.[a]

	Average	GM[b]	GSD[c]	Range
Radon gas concentration (Bq m^{-3})	177	142	2.02	32–492
Glass age (y)	20	19	1.5	7–50
Radon gas exposure[d] (kBq y m^{-3})	4.3	2.6	2.3	0.75–38

[a] Samples from site SD not included.
[b] Geometric mean.
[c] Geometric standard deviation.
[d] Includes imputed values.

Comparison of surface activity detectors

The most common surfaces were glass that covered photographs or paintings (54%), mirrors (23%), and windows (13%). Most houses had two sampling sites in separate rooms. The surfaces do not represent a random sample, even among the houses surveyed. In houses with cooperative occupants, every useful glass surface in a living space near a long-term radon gas measurement site was sampled. Twenty-one of the sixty-nine samples came from one house with an almost complete radon gas history. However, subset analyses show that the inclusion or exclusion of this house had little effect on the detector performance intercomparison or the exposure-activity relationships. Approximately 19% of the glass surfaces were exposed to one or more pack-years of tobacco smoke. Table 2 summarizes the distribution of the exposure data.

Both detectors performed well in the laboratory and field quality assurance tests. In side-by-side placements, the coefficient of variation (CV) was 6% for the RRDs and 14% for the RSMs. Sequential measurements on the same area of the same glass also yielded reproducible results: 10% for the RSM and 14% for the RRD. The RRDs that were exposed for 1 y showed the same activity as the RRDs exposed for 60 d ($R^2 = 0.93$, slope = 1.01). Lab blanks had few tracks. Field blanks had more tracks than were anticipated. The exposures were equivalent to a corrected surface activity of 0.3 Bq m^{-2} for the RRD and 0.5 Bq m^{-2} for the RSM. A test measurement on a glass sample that had only been exposed for 1 mo in house SD suggested that the natural alpha-emitting contamination was not being completely eliminated by either detector's reduction technique. The equivalent activity, had the glass been believed to be 20 y old, would have been 1.0 Bq m^{-2} for the RRD and 3.0 Bq m^{-2} for the RSM. In response to that observation, RRDs were placed on 11 different new glasses for a 206-d exposure in the lab. These exposures showed an average contamination rate equivalent to 1.3 Bq m^{-2} with a standard deviation of

Table 2. Radon gas and surface activity distributions by detector type.

	RRD[a]	RSM[b]
Number of buildings	24	25
Number of surfaces	67	65
Radon gas exposure (kBq y m^{-3})	2.21 (2.14) 3.49[c]	2.22 (2.13) 3.50[c]
Surface activity (Bq m^{-2})	5.52 (2.14) 7.87[c]	9.11 (1.88) 11.40[c]

[a] Retrospective reconstruction detector used in the Iowa radon lung cancer study.
[b] Retrospective surface monitor used in the Missouri radon lung cancer study.
[c] Geometric mean (geometric standard deviation) average values.

1.0 Bq m^{-2}. One RSM placed side-by-side on an unexposed lab sample reported an equivalent activity approximately twice the RRD activity. Although more work needs to be done, it appears that neither detector's contamination reduction protocol is optimized.

When exposed side by side on household glass, the RSM and RRD report activities that are highly correlated (R^2 ~0.9). Table 2 shows the exposure and activity distributions for each detector. Although the exposure conditions are similar, the activity distributions are slightly different. Both paired *t* tests and Wilcoxon signed rank tests suggest that the RSM and RRD results are different. Linear regression analysis of the two detector's activity measurements suggests that the slope and intercept are strongly correlated. The slope is near one and the intercept is near zero (Fig. 1). A comparison of the two detectors' track generation rates, rather than corrected activities, shows a similar pattern of slight disagreement. This suggests that the systematic difference probably comes from the different methods used to subtract contamination tracks. The RSM disagreed by a factor of ten with the RRD and a factor of five with the cumulative radon gas exposure on two window surfaces in a single room (Figs. 1 and 2). While it is likely that the RSMs from those windows developed additional tracks in transport or storage, we had no solid evidence to exclude them based on that suspicion. The inclusion of those two RSM data points did not significantly alter the average results of the analyses.

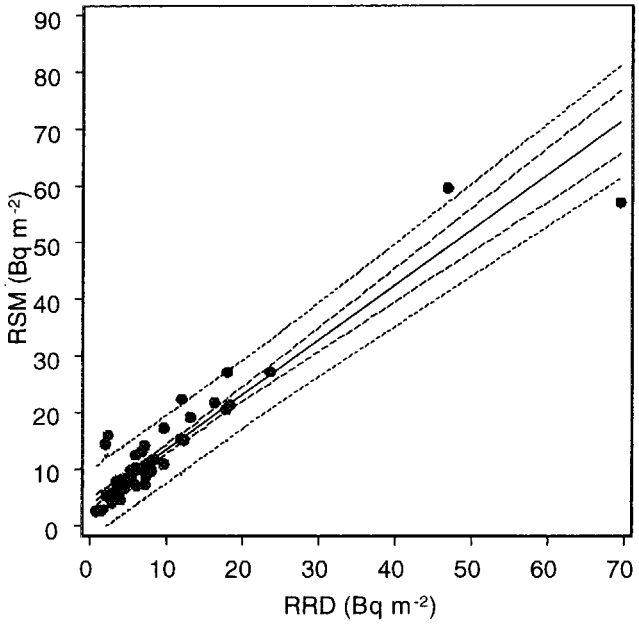

Fig. 1. Intercomparison of two types (RRD and RSM) of retrospective radon gas detector activities: The detectors were exposed side-by-side. The central solid line is the regression fit. The pair of dashed lines closest to the fit line represents the 95% confidence interval of the fit. The pair of dotted lines represents the 95% confidence interval of the data.

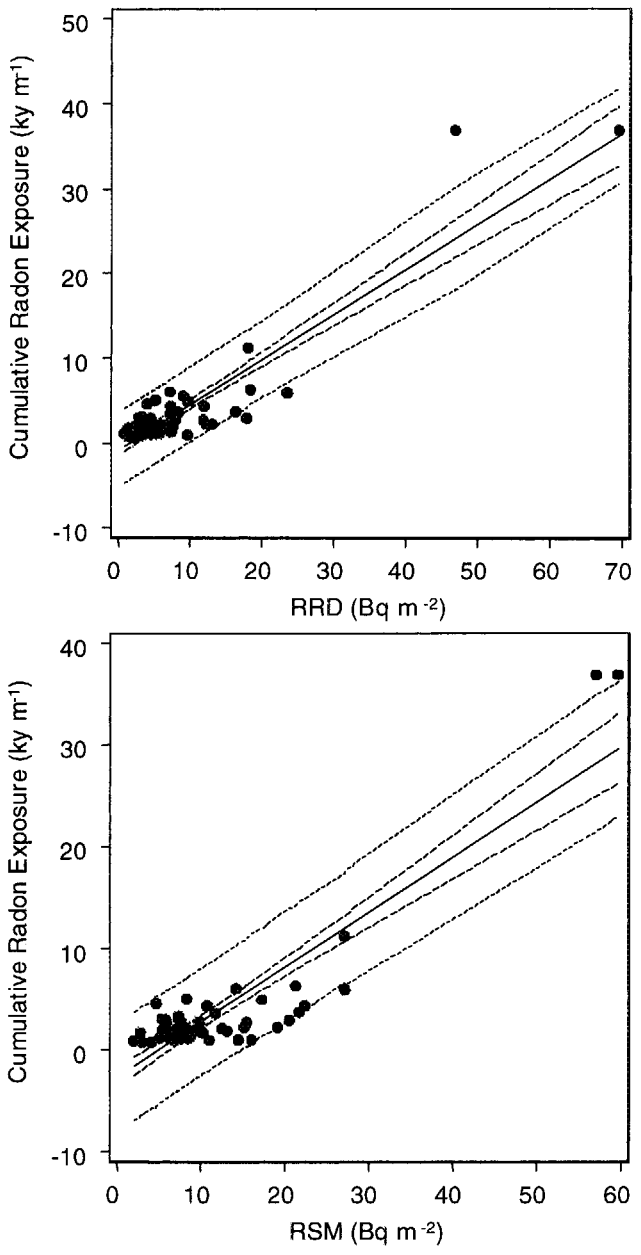

Fig. 2. Radon gas exposure and adjusted implanted activity for two types of retrospective radon gas detectors (a) RRD and (b) RSM: The central solid line is the regression fit. The pair of dashed lines closest to the fit line represents the 95% confidence interval of the fit. The pair of dotted lines represents the 95% confidence interval of the data.

Analysis of radon gas exposure vs. surface activity

Linear regression analysis showed good correlation (R^2 ~0.8–0.85) between the cumulative radon gas exposure (kBq y m^{-3}) and the surface activity (Bq m^{-2}) for

both types of detectors (Table 3). Surface activity measurements produced cumulative radon gas exposure estimates that were accurate to within 25%, on average. The slopes of the RRD and RSM exposure-activity regression lines do not differ significantly; both are approximately 0.57 ± 0.03 ky m^{-1}. The intercepts do differ, most likely due to the difference in subtracting contamination contributions. Table 3 shows that no significant changes in regression parameters occur when some categories of samples (smoke exposed, windows, house SD) are excluded from the analysis. Unfortunately, the small number of samples in these categories limits the ability of the analysis to detect categorical differences. No substantial change in fit parameters or coefficients of determination occurred when the analyses used only the most reliably imputed cumulative exposures.

Comparison of retrospective radon gas reconstruction methods

A surface activity measurement (RRD-RSM) had about the same accuracy in retrospectively reconstructing the cumulative radon gas exposure as a single yearlong radon gas measurement (ATD). That is, in the random sampling simulation, the confidence intervals of the coefficient of determination distributions for the ATD and RRD-RSM estimators overlapped. For the site-based simulation, the ATD coefficient of determination confidence intervals ranged from 0.84 to 0.95 while the RRD-RSM interval ranged from 0.80 to 0.86. The comparable ranges in the house-averaged simulation were 0.85 to 0.95 for both the ATD and the RRD-RSM exposure estimators. Thus, either estimator is within about 20 to 25% of the actual cumulative radon gas exposure.

DISCUSSION

The good agreement between the surface activity measurements from two different detectors and the good correlation between the measured cumulative radon gas exposure and the measured surface activity provide new support for the use of glass-implanted ^{210}Po to estimate past radon gas exposure. The slope of the exposure-activity regression line in the present study is similar to the results for the RRD and RSM in earlier studies (Mahaffey et al. 1993; Lively and Steck 1993; Field et al. 1999; Steck and Field 1999a and b; Mahaffey et al. 1999). We expect some differences in these slopes since the studies took place in houses with different depositional environments. For example, the number of smokers' houses is much higher in the epidemiologic study groups. The coefficients of determination of the current study are substantially higher than our previous studies (Lively and Steck 1993; Mahaffey et al. 1993; Field et al. 1999; Mahaffey et al. 1999; Steck and Field 1999a and b). Actual radon measurements cover most of the glass exposure period in the present study since the annual average radon gas concentration has been measured repeatedly over a 10- to 15-y period while earlier investigations relied on a single contemporary radon gas measurement to estimate past radon concentrations. Although our sample size is small, the stability of the fit parameters in the sub-groups analyzed suggests that glass activity measurements are useful for assessing cumulative radon exposure.

Our fit results are similar to a recent prospective exposure study where new, identical glass surfaces were exposed in high radon homes concurrently with radon gas measurements (Fitzgerald and Hopke 2000). The coefficient of determination in that study increased from 0.81 to 0.99 when homes that had high or low deposition rates were eliminated. Earlier work suggests that the fit to our data may improve if the implanted activity is adjusted by using the activity measured by a separate deposition chip on the RRD to account for different depositional environments (Steck and Field 1999a and b).

Our study is limited by the number and variety of surfaces studied. In particular, only 19% of the glass was

Table 3. Regression analysis of cumulative radon gas exposure as a function of surface activity.

	RRD				RSM			
	N	Slope[a]	Intercept[b]	R^2	N	Slope[a]	Intercept[b]	R^2
All samples	67	0.56 ± 0.03	−0.9 ± 0.4	0.85	65	0.56 ± 0.03	−2.8 ± 0.5	0.81
Non-smoking[c]	53	0.57 ± 0.03	−1.3 ± 0.4	0.87	51	0.58 ± 0.04	−3.6 ± 0.6	0.84
Non-windows	57	0.56 ± 0.03	−0.8 ± 0.4	0.86	58	0.57 ± 0.03	−2.6 ± 0.5	0.85
Non-SD[d]	44	0.57 ± 0.03	−0.8 ± 0.3	0.86	45	0.57 ± 0.04	−2.9 ± 0.4	0.83
Best exposure	57	0.56 ± 0.03	−1.0 ± 0.4	0.86	55	0.57 ± 0.04	−2.8 ± 0.4	0.83

[a] Linear regression of exposure and activity; units are ky m^{-1}, equivalent to kBq y m^{-3} divided by Bq m^{-2}.
[b] Units are Bq m^{-2}.
[c] Nonsmoking glass is defined as having been exposed to no more than one pack year of environmental tobacco smoking by occupants of the house.
[d] Samples from site SD not included.

subjected to cigarette smoke while we estimate that, in the Iowa study, 80% of the glass was exposed to smoke. In addition, the glass in the present study was exposed primarily in one house with constant adult occupants, which is a situation similar to the Iowa study, but not in most of the published epidemiological studies.

What makes a better retrospective radon gas meter, yearlong ATD radon gas measurements or RRD-RSM surface activity measurements? Currently yearlong radon gas measurements are considered the best-available radon gas measurement standard for homes. Yearlong measurements can suffer from year-to-year radon gas variations. Surface activity measurements may suffer from the variations in the deposition environment, long-term loss of activity, and impediments to implantation (Cauwels and Poffijn 2000; Fitzgerald and Hopke 2000; Fleischer and Doremus 2001). However, little is known about the actual magnitude of these potential variations. The random sampling simulation produced similar coefficient of determination distributions for yearlong gas and surface activity measurements. Thus, whatever environmental variations affect the surface measurements, they introduce about as much uncertainty in the cumulative radon exposure estimate as the year-to-year variation uncertainty introduces in yearlong radon gas measurement-based estimates. Of course, we must keep in mind that these data are not from a widely divergent set of conditions but rather represent a baseline study done in houses (mostly non smokers) with constant occupants and well-known radon histories.

Since surface activities reflect radon progeny deposition, the surface activity measurements may be used to measure airborne radon progeny dose. We tested this idea in a Monte Carlo simulation of the airborne radon-related dose rate and surface deposited activity in a sample of houses with the characteristics of those in the Iowa study. This semi-empirical model calculates the fate and transport of the radon progeny aerosols using environmental parameter distributions (e.g., air exchange rates, number of smokers, etc.) that are representative of a specific group of houses. The details of the model are given elsewhere (Lively and Steck 1993; Steck and Field 1999a and b; Steck 2002). In this simulation, we held the radon gas constant across houses so that the simulation was sensitive to the dose variations due to environmental factors alone. Although the loss of activity to the walls might lead one to expect a negative correlation between airborne dose rate and surface activity, the surface activities actually showed a positive correlation with the airborne dose rate in these houses. The correlation results from the highly mobile radon progeny nucleation mode, which has a high deposition rate and a high dose conversion factor. Thus, surface activity measurements, coupled with radon gas measurements, may yield a better dose estimate than radon gas measurements alone.

One of the major limitations of past residential epidemiologic studies was the loss of power from the inability to estimate radon gas concentrations accurately when subjects occupied numerous homes (Lubin et al. 1990; Weinberg et al. 1996). Some studies, like the Iowa Radon Lung Cancer Study, overcame this limitation by limiting enrollment to subjects who spent at least 20 years in their current home (Field et al. 2000). The Missouri Radon Lung Cancer Study, which utilized both radon gas measurements and surface activity measurements to estimate historical radon gas exposure, did not limit their enrollment to subjects with extended occupancy in the current home (Alavanja et al. 1999). The Missouri study found a statistically significant lung cancer risk when the analysis incorporated surface activity measurements, but not for the analysis that relied on radon gas measurements. The good correlation between cumulative radon exposure and surface activity suggests that surface activity measurements may have decreased exposure misclassification and increased the power of the Missouri study to detect an association between prolonged radon exposure and lung cancer by providing a better estimate of the retrospective radon gas exposure for items that traveled along with the homeowners as they moved from one home to another. Surface activity measurements also allow pooling of results from two studies, like the Iowa and Missouri studies, even though their minimal residency requirement for participants differed.

Finally, surface activities have another advantage for applications that require rapid, yet accurate results. Implanted surface activities can be measured by a variety of techniques in periods much shorter than a year. Inexpensive track registration detectors have been shown to produce accurate cumulative radon gas exposure results after a 60-d exposure. Large area semiconductor diode detectors can measure surface activity even more rapidly. For example, using a 20-cm^2-semiconductor diode detector, it would only take 1 d to measure a piece of glass that had been exposed for 10 y in a 150 Bq m^{-3} house.

CONCLUSION

The present study provides new support for the use of implanted surface activity to estimate cumulative radon gas exposure. Glass surface activity measured by two different types of detectors was able to estimate the long-term average radon gas concentration as accurately as yearlong radon gas measurements. Surface activity measurements can provide reliable estimates of past

radon gas concentrations for rapid home screening tests. They are also useful in retrospective epidemiologic studies that include participants who may have been exposed to highly variable radon concentrations in previously occupied or structurally modified homes. Further refinements of these glass-based retrospective detectors and calibration in a wider variety of depositional environments may improve their ability to reconstruct cumulative radon gas and radon-related dose estimates.

Acknowledgments—We thank Joan Huber, Survey Research Associates for assistance with data collection, and Cheryl Smith of Dade Moeller & Associates for analysis of the RSM glass detectors. This publication was made possible by contract number 263-MQ-820009 and grant number R01 CA859422 from the National Cancer Institute, National Institutes of Health. This report is solely the responsibility of the authors and does not necessarily reflect the official views of the NCI or NIH.

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